



# Study of the effects of regeneration of USY zeolite on the catalytic cracking of polyethylene

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## ABSTRACT

In this work, a study of the deactivation and reuse of USY zeolite for the pyrolysis of polyethylene has been carried out in a batch reactor at 500 °C. From the different tested zeolite/plastic ratios, 1/10 is the optimal ratio which allows giving rise to a liquid product free of wax. Regeneration was established by heating at 500 °C for 3 h coked zeolite in presence of air. The liquid yield begins to increase from the 11<sup>th</sup> cycle of regeneration until reaching a maximum at the 14th cycle showing a lost in the zeolite activity producing a viscous product.

The liquid fraction derived from the first eight cycles was a mixture of C<sub>4</sub>–C<sub>27</sub> compounds with a maximum C<sub>5</sub>–C<sub>7</sub> fraction. In the 14th cycle of regeneration, compounds having higher carbon number were obtained and the yield of linear compounds increases up to 29%. To explain the relative activity of the regenerated zeolite, the textural properties and the XRD patterns results of the zeolite recovered after the 1st and 14th cycles have been compared with those obtained with fresh catalyst. The study showed that after 14 cycles of regeneration, the zeolite is still active but its activity level has decreased.

## 1. Introduction

Continuous consumption of plastics led to the accumulation until 2015 of 6.3 billion tons of plastic waste, of which only 30% were recycled [1]. Hence, the recovery of this waste by pyrolysis can be considered as an appropriate method against incineration and landfilling.

Polyethylene (PE) is the main component of plastic waste [2] and its thermal degradation favors the formation of wax containing a large amount of alkanes in a wide range of molar masses [3]. Therefore, the use of catalysts gives the pyrolysis an added value, since a suitable catalyst can improve product quality and the process lead to a good selectivity to more valuable products, even at lower temperatures than thermal pyrolysis [4].

Because of their porous structure and acidic properties, zeolitic (ZSM-5 [5], Y-zeolite [6]) and mesoporous materials [7] are the most important catalysts used in catalytic cracking of polymers. The Ultra-stable Y (USY) zeolite, having large pore size tends to produce more light hydrocarbons, mainly in the range of gasoline [8]. However, during the catalytic degradation of plastics in the presence of catalysts, especially zeolites, coke deposits are formed, covering the active sites and blocking the pores of the zeolite, which leads to its deactivation in

short periods [9].

The options available for treating deactivated catalysts are elimination or regeneration. Disposal is undesirable from an economic and environmental point of view as it leads to the loss of expensive metals, while environmental laws for the disposal of potentially hazardous materials have become more stringent [10]. In addition, USY zeolite is an expensive catalyst and its use in large amounts in an industrial plant could affect the economy of the process.

Therefore, regeneration is the most desirable alternative and involves the treatment of the catalyst in order to recover and / or restore the lost catalytic activity in order to extend its life cycle and delay its elimination.

Few studies are available in the literature about USY deactivation and regeneration in pyrolysis of polyethylene and no studies have been conducted yet to show the effects of different regeneration cycles on the liquid produced during catalytic cracking of polyethylene.

Marcilla et al. [11] have studied the deactivation behavior of HZSM-5 and HUSY during the catalytic pyrolysis of polyethylene using a thermogravimetric analysis (TGA). They reported that HUSY zeolite presents a fast deactivation compared to the small pore size in HZSM-5 which prevents coke deposition.

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Lin et al. [12] studied the deactivation of USY zeolite during the catalytic pyrolysis of high density polyethylene using TGA analysis. They found that the catalyst activity decreased exponentially with the coke content and zeolite regained most of initial activity after regeneration. The effect of the final cracking temperature on the regeneration behavior and the location of the coke obtained in the catalytic pyrolysis of LDPE in the presence of HZSM-5 and USY zeolites have been studied by Marcilla et al. [13] using a TGA analysis. They reported that the combustion of HZSM-5 coked samples occurs at higher temperatures as compared with those obtained with USY zeolite.

The objective of this study is to investigate the ability of the zeolite USY to remain active during several regeneration cycles and to be used several times in the pyrolysis of polyethylene until its exhaustion in a lab-scale reactor. For this, first the amount of zeolite to adopt during the experimental tests was optimized. Then, the influence of regenerated USY zeolite on the yield and composition of the derived oils from catalytic cracking of PE was studied. Finally, different analytical techniques have been carried out in order to characterize the fresh and coked zeolites and study the catalyst deactivation.

The originality of this work resides on the investigation of the effects of different regeneration cycle of USY zeolite on both liquid products distribution and catalyst characterization which has never been done before.

## 2. Experimental

### 2.1. Materials

PE samples were gathered from municipal waste collection facility. Feedstock was washed, dried and crushed into small pieces (2.2–4 mm) in order to get a homogeneous mixture. The elemental analysis of polyethylene samples showed an H/C mass ratio of 16.73% (14.43/86.23) and no heteroatoms were detected.

In addition, no deposit of ash was recovered after heating the material in a muffle furnace at a temperature of 800 °C, which indicates that the plastic additives amounts are negligible.

A commercial USY zeolite purchased from TOSOH Corporation was used for the catalytic experiments. Its main characteristics are presented in Table 1. In order to remove moisture content, the catalyst was dried in an oven, and then was sieved in order to have a particle size between 200 and 800 µm.

### 2.2. Cracking experiments

The pyrolysis experiments were carried out using a batch reactor, developed by Pyrum Innovations, as described in our previous work [14]. In a typical run, 200 g of polyethylene mixed with 20 g of USY zeolite were placed into the reactor which was heated to 500 °C. The pyrolysis products were cooled using a condenser fed with tap water at 15 °C and the condensate was collected in a container. The uncondensed gases were discharged through an exhaust system.

The regeneration of the USY zeolite was carried out in a muffle furnace in presence of air. The temperature program includes a ramp of 10 °C.min<sup>-1</sup> up to 500 °C and then maintained for 3 h.

**Table 1**  
Textural and acid properties of the fresh USY zeolite.

SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> (mol/mol)	15
BET surface area (m <sup>2</sup> g <sup>-1</sup> )	700
t-Plot External surface area (m <sup>2</sup> g <sup>-1</sup> )	106.26
Na <sub>2</sub> O (wt%)	0.05
Micropore volume (MPV) (cm <sup>3</sup> g <sup>-1</sup> )	0.255
Crystal size (µm)	0.7–1.0
Total acidity (mmol NH <sub>3</sub> g <sup>-1</sup> )	0.1

### 2.3. Catalyst characterization

#### 2.3.1. Specific surfaces measurements

The measurement of specific surfaces, microporous and mesoporous volumes of the fresh and coked samples and the adsorption isotherms was carried out by nitrogen adsorption/desorption at 77 K using a Micromeritics ASAP 2020 volumetric apparatus.

Previously, the samples were outgassed under vacuum (about 40 mmHg) in order to eliminate most adsorbed impurities (water in particular) and heated from room temperature to 350 °C with a heating rate of 10 °C/min for at least 48 h. Surface areas were calculated by application of the BET equation. Micropore volume and external surface area of the catalysts were obtained by using the t-plot method.

#### 2.3.2. Crystalline structure

XRD (X-Ray Diffraction) measurements were performed using a SIEMENS Diffraktometer D5000 using Cu Kα radiation generated at a voltage of 40 kV and a current of 40 mA with a pitch of 0.02. The scanning range of 2θ was 5° to 70° with a scan rate of 2 s.

#### 2.3.3. Acidity characterization by adsorption of pyridine followed by infrared analysis

Samples of 20–30 mg of fresh and regenerated zeolite (after 1 and 14 cycles) were pelletized and treated overnight at 450 °C under air flux, then they were cooled down until 200 °C. A secondary vacuum was then generated in the analysis cell during 1 h. A Thermo Nicolet 6700 spectrometer was used at these conditions, in order to register the reference spectrum between 1454 and 1545 cm<sup>-1</sup>. Then the cell was cooled down until 150 °C where the Pyridine was absorbed. Secondary vacuum was applied again before collecting the new spectrum. The desorption by temperature increase up to 450 °C was then realized. The infrared band at 1454 cm<sup>-1</sup> area was measured at different temperatures in order to measure Lewis acid sites while that of the band at 1545 cm<sup>-1</sup> was used in order to measure Bronsted acid sites. Spectrum at 150 °C was used to determine the number of acid sites while that of 350 °C indicates the strength of these sites.

### 2.4. Liquid products characterization

In order to study the difference of produced oils components carbon number distribution of from different pyrolysis reaction in presence of the regenerated catalyst, analyses were performed using a PERKIN ELMER Clarus 680, coupled with a Perkin Elmer mass spectrometer and a Flame Ionisation Detector (GC/MS-FID). An Agilent SLB-5MS column (30 m × 0.250 mm; 0.25 µm film thickness) was used with a helium flow rate of 1 ml/min. The temperature program began with a hold at 40 °C for 10 min followed by an increase at 3 °C/min to 170 °C, then 10 °C/min to 340 °C, which was held for 6 min. The temperature of FID detector was fixed at 250 °C. Injections of 1 µl of oil samples were realized with a split ratio of 40:1. The mass spectrometer was set at an ionizing voltage of 70 eV and a range of m/z 30–450. Identification of chromatographic peaks was established from the NIST library.

## 3. Results and discussion

### 3.1. Optimization of zeolite/plastic ratio

Previous works have shown that the non-catalytic pyrolysis of PE produces 80% condensable fraction with high wax proportion and 20% gas [15]. For this, the study of the optimization of the catalyst/plastics ratio was carried out in order to obtain a liquid as a product. For this, different zeolite/plastic ratios were tested with PE: 0.5/10, 0.8/10 and 1/10. The optimal ratio is the one which allows giving rise to a liquid product free of wax.

From Fig. 1, it can be seen that by reducing the zeolite content by up to 5%, the produced liquid contained up to 30% wax (corresponding to

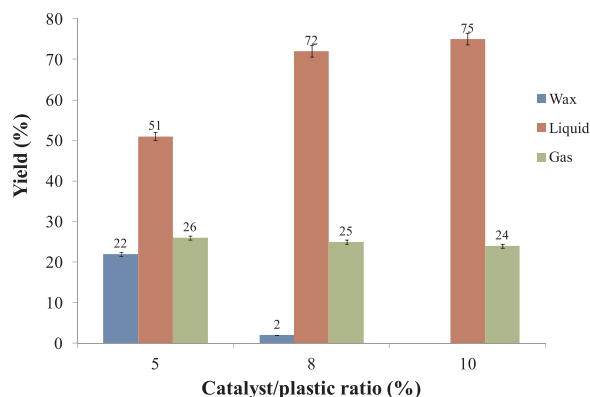


Fig. 1. Yield of products according to zeolite content.

22% of the plastic mass). An increase of up to 8% leads to a slight wax content. So, to ensure a good operating system during experiments, the zeolite/plastic ratio of 1/10 was adopted.

### 3.2. Effect of zeolite regeneration on the yield of the liquid product

In order to study the effects of regeneration on the activity of the USY zeolite, the latter has been used in the consecutive cracking reactions of PE and regeneration cycles until its exhaustion which is manifested by a degradation of the quality of the liquid and the appearance of wax in the pyrolysis products. The first criterion studied is the evolution of the yield of the condensable and gaseous fractions over the regeneration cycles.

Fig. 2 represents the relative difference in the yield of condensable and gaseous fractions between two consecutive regeneration cycles

As it can be seen, the USY zeolite maintained an almost stable behavior during the first 8 cycles of regeneration showing the efficiency of the regeneration process. Whereas, from the 10th cycle, the yield of condensable fraction begins to increase until reaching a maximum at the 14th cycle. A simultaneous decrease in gas yield was observed on the counterpart. This shows that the zeolite begins to lose its activity especially that the product obtained in the last cycle is more viscous than the one before. For each reaction cycle, the acidity of the catalyst decays, resulting in a decrease in the polymer cracking activity and a lower production of low molecular weight components which are collected as a gas. In fact, the deactivated catalyst becomes less effective in polymer cracking. The lower activity of the deactivated catalyst results in lower cracking activity compared to the virgin catalyst which leads to heavier products. This is manifested by higher production of condensable products but having higher content of heavier hydrocarbons.

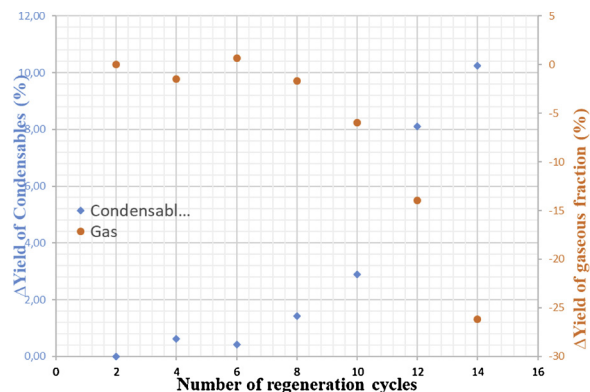


Fig. 2. Relative evolution of condensable and gaseous fractions yields as a function of regeneration cycles (standard error for yield is 2%).

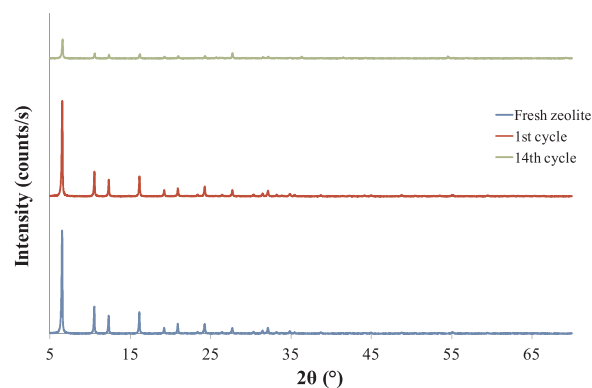


Fig. 3. XRD patterns of the fresh and recovered zeolites after the 1st and the 14th regeneration cycle.

### 3.3. Characteristics of the fresh and coked zeolites recovered at different regeneration cycles

#### 3.3.1. XRD analysis

XRD study was carried out in order to verify the crystalline structure of the fresh and the recovered zeolites after the 1st and 14th regeneration cycles. Fig. 3 shows the variation of crystallinity as a function of the regeneration cycles.

It can be seen from the diffraction patterns obtained for the fresh zeolite and that of the zeolite recovered after the 1st and the 14th cycle that the regeneration did not change the crystalline structure because all the samples presented typical peaks and have retained the topological form of a FAU zeolite [16], however, the signal strength decrease with respect to cycles indicates that the crystalline structure has been damaged. This damage could be caused essentially by the exothermal reaction of coke oxidation that creates hot spots inside the pores of zeolite and breaks the Aluminosilicate framework leading to dealumination [17] and extra-framework species apparition [18].

#### 3.3.2. Acidity characterization

In order to investigate this possibility, the results of acidity characterization were analyzed and presented in Table 2. It can be seen that Lewis acid sites are increasing with regeneration cycles until almost reaching the double at the 14th cycle. At the same time, Bronsted sites concentration slightly increased after 1st regeneration cycle and drastically decreased after the 14th cycle to reach 20% of its initial value. The strengths of both types of acid sites have slightly increased after one cycle and drastically decreased after the 14th.

Usually, Lewis acid sites are caused by  $Al^{3+}$  presence that is usually caused extraframework aluminum [19,20]. Bronsted sites are usually caused by Si–OH–Al bonds inside the tetrahedral clusters. Extra-framework aluminum could enhance the stability and strengths of Bronsted acid sites because it equilibrates charges of clusters when it is blocked inside the supercage of Faujasite. This effect is also intentionally provoked during the dealumination of Faujasites [18]. However, this effect disappears when the zeolite lattice is damaged and Bronsted sites decrease and weakens with the increase of amorphous aspect of material. From XRD results, it could be concluded that the extraframework aluminum lead to increasing sites concentration and strength because the material is still highly crystalline and thus extra-framework species are more likely to be trapped inside the crystalline structure. On the other hand, the loss of crystallinity at the 14th cycle leads to weak Bronsted acid densities and strengths.

#### 3.3.3. BET analysis

As can be seen in Table 3, the BET surface and the pore volume of the zeolite recovered after the 1<sup>st</sup> regeneration cycle have slightly decreased compared to those of the fresh zeolite, which indicates that the

**Table 2**  
Acidity analysis of fresh and regenerated zeolites.

	Fresh zeolite	1st Regeneration	14th Regeneration
Lewis sites Density <sup>a</sup> (μmol/g)	46	60	89
Lewis sites strength <sup>b</sup> (μmol/g)	15	23	4
Bronsted sites Density <sup>a</sup> (μmol/g)	41	47	8
Bronsted sites strength <sup>b</sup> (μmol/g)	17	18	0

<sup>a</sup> measured at 150 °C.

<sup>b</sup> measured at 350 °C.

**Table 3**  
texture characteristics of the fresh and recovered zeolite after the 1st and 14th regeneration cycles.

	BET Surface (m <sup>2</sup> /g)	t-Plot External Surface Area (m <sup>2</sup> /g)	Pore volume (cm <sup>3</sup> /g)	t-Plot Micropore volume (cm <sup>3</sup> /g)
Fresh zeolite	700	110.01	0.466	0.255
1 <sup>st</sup> cycle	690	106.26	0.450	0.249
14 <sup>th</sup> cycle	387	64.95	0.274	0.139

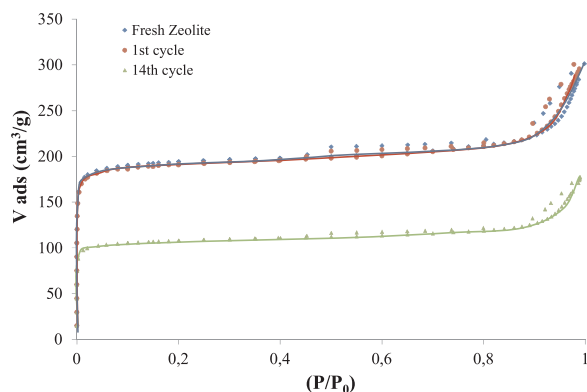
zeolite is still active. After 14 cycles of regeneration, the BET surface area decreased by about 45%, which is indicative of the destruction of crystalline structure and the deposition of extraframework species leading to the blocking of the pores.

The nitrogen adsorption and desorption isotherms obtained for the fresh zeolite and the zeolites recovered after the 1<sup>st</sup> and 14<sup>th</sup> regeneration cycles are presented in Fig. 4.

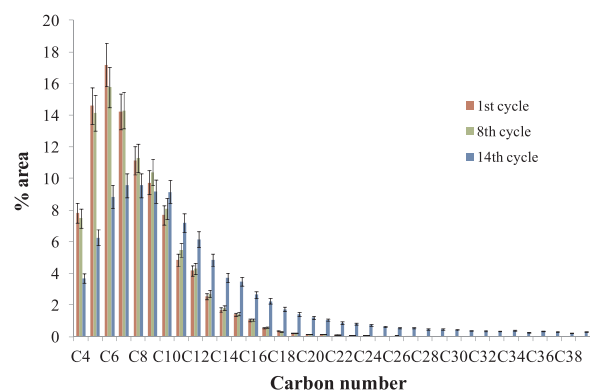
The isotherms represented correspond to a type I zeolite in the classification of (Brunauer et al., 1938) and corresponds to a solid microporous.

The zeolite is characterized by a tray for the initial part ranging from very low values of P/P<sub>0</sub> and for the final part; there is an increase of the slope that reveals a hysteresis phenomenon characteristic of mesoporous medium [21].

The isotherms of the fresh zeolite and the zeolite recovered after the 1st regeneration cycle are very close, which shows that the zeolite has retained its properties after the first regeneration. The slight decrease in BET area and in micropores volume could be attributed to the deposition of small amounts of extraframework species in the pores of the zeolite. While the isotherm of the zeolite recovered after the 14<sup>th</sup> regeneration cycle still has the same aspect but with a much lower adsorption volume than that of the fresh zeolite which indicates the destruction of crystalline structure and the blockage of pores by extraframework species.



**Fig. 4.** Nitrogen adsorption-desorption isotherms of fresh and recovered zeolites after the 1<sup>st</sup> and 14th regeneration cycle.



**Fig. 5.** carbon number distribution of the liquid derived from the catalytic pyrolysis of PE during the 1st, 8th and 14th cycles.

### 3.4. Effect of regeneration of zeolite on the chemical composition of the liquid fraction

The carbon chain length distributions of the 1st, 8th and 14th regeneration cycle liquid products are illustrated in Fig. 5.

It can be seen that the liquid obtained from the catalytic pyrolysis of PE in the first 10 cycles is rich in hydrocarbons whose carbon number vary between C<sub>4</sub> and C<sub>27</sub> with a maximum C<sub>5</sub>-C<sub>7</sub> fraction. After the 10th cycle, gradual changes in liquid products appeared and the carbon number range starts to expand until reaching C<sub>39</sub> at the 14th one. This expansion of carbon chain range was accompanied with a considerable decrease in the proportion of the C<sub>4</sub>-C<sub>6</sub> fraction all along with a simultaneous increase of heavier compounds. Wax that is represented with carbon chains longer than 20 carbon atoms increases slowly at the first 10 regenerations and sharply at the last 4 ones where it reaches 21.4% of the total condensable fraction.

Further analysis of liquid products during regeneration cycles such as yield of linear compounds (paraffin/olefin) and yield of branched and aromatic compounds was also established (Table 4).

From Table 4, the results clearly show how the zeolite is being deactivated. Catalytic pyrolysis in the presence of the fresh zeolite produced only branched compounds. It may be noted that during the first 8 regeneration cycles, small changes in the yield of the linear and branched compounds, and therefore, the zeolite is still active and still has the potential to be used in the pyrolysis of the polymer. These observations are emphasized with Bronsted acidity strength and density decrease at the 14th cycle. It is generally known that catalytic cracking of hydrocarbons depends on the concentration and the strength of Bronsted sites [18].

In fact, the yield of condensables in thermal pyrolysis of polyethylene is around 80% containing large proportion of heavy linear hydrocarbons, with a high carbon chain number [14].

In presence of acidic catalyst as USY zeolite, PE cracking is known to proceed by carbocation mechanisms, where the initially formed ions undergo chain reactions via processes, such as scission or β-scission and isomerisation and hydrogen transfer alkylation and oligomerisation, to yield cracked products having lower molecular weights [22]. The acid



**Table 4**

Composition of the liquid obtained at the different regeneration cycles.

Regeneration cycles	No Catalyst	Fresh catalyst	2	4	6	8	10	12	14
Proportion of wax ( $C_{>20}$ ) in liquid (%)	52	0.98	1.03	1.59	1.65	1.8	2.98	10.11	21.42
Proportion of linear compounds (%)	75	0	6	8	8	8	8	17	29
Proportion of branched + aromatics compounds (%)	25	100	94	92	92	92	92	83	71

strength and density have a significant impact on the catalytic activity of Y zeolites. The bonds of the heavy components can be polarized and cracked in the presence of proton which is generated from Bronsted acid sites, and then converted into light aromatic and branched products [23]. When deactivation occurs, the catalyst is not efficient for cracking waxes, which are the main products obtained at thermal pyrolysis. For this, the formation of wax and the change in product distribution, are an index of catalyst deactivation [24].

It can be seen that from the 10th regeneration cycle, the yield of compounds having a carbon number greater than  $C_{20}$  begins to increase until reaching 10% in the 12th cycle and 21% in the 14th cycle. Similarly, the yield of linear compounds increases up to 29% at the 14th cycle, which reflects that the zeolite is still active but its activity level has decreased due to the decreased number and strength of Bronsted sites

As USY zeolite has tridimensional pore structures, it is capable of trapping the molecules [17]. The limitation of access of the reactant to the active sites or the blockage of the access can lead to the deactivation of USY zeolite catalyst [25]. This blockage could be a result of crystalline structure destruction and the formation of extra-framework species that would obstruct faujasites cavities. This aspect can be seen obviously from XRD, BET and FTIR results (cf. section 3.3)

This loss of zeolite activity could be compensated by increasing the proportions of regenerated catalyst in the plastic charge or by mixing fresh zeolites with regenerated ones (aspects that were not covered in this paper).

#### 4. Conclusions

In this work, the effects of regeneration of USY zeolite on the cracking of polyethylene have been performed using a batch reactor.

USY zeolite showed a stabilization behavior during the first 8 cycles of regeneration showing the efficiency of the regeneration. While from the 10th cycle, the zeolite begins to lose its activity and at the 14th cycle, wax started to appear in the product. This decrease of reactivity could be overbalanced by increasing the catalyst/plastic ratio.

Based on the GC analysis, the liquid obtained from the catalytic pyrolysis of PE in the 10 first cycles is rich in branched and aromatic compounds whose carbon number varies between  $C_4$  and  $C_{27}$  with a maximum  $C_5$ – $C_7$  fraction. During the 14th cycle, the liquid was distributed over a wide range of carbon numbers ( $C_4$ – $C_{39}$ ) containing 29% of linear compounds.

Based on the BET and XRD analysis, the regeneration gradually broke the crystalline structure. On the other hand, the intensity of the peaks and the isotherms has decreased significantly between the fresh zeolite and the zeolite recovered after the 14th cycle, showing a decrease in the crystallinity. FTIR results after desorption of pyridine showed that the destruction of crystalline structure has increased Lewis sites density. While Bronsted sites density has increased after one regeneration cycle and then decreased 5 times at the 14th regeneration cycle.

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